Superconductivity in SrNi$_2$As$_2$ Single Crystals

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(Dated: August 12, 2008)

Abstract

The electrical resistivity $\rho(T)$ and heat capacity $C(T)$ on single crystals of SrNi$_2$As$_2$ and EuNi$_2$As$_2$ are reported. While there is no evidence for a structural transition in either compound, SrNi$_2$As$_2$ is found to be a bulk superconductor at $T_c = 0.62$ K with a Sommerfeld coefficient of $\gamma=$ 8.7 mJ/mol K^2 and a small upper critical field $H_{c2} \sim 200$ Oe. No superconductivity was found in EuNi$_2$As$_2$ above 0.4 K, but anomalies in ρ and C reveal that magnetic order associated with the Eu^{2+} magnetic moments occurs at T_m=14 K.

PACS numbers: 74.10.+v,74.25.Bt,74.70.Dd

The ThCr$_2$Si$_2$ structure type is well known for accommodating a superconducting ground state, particularly in the heavy fermion community with superconductors such as CeCu$_2$Si$_2$ and URu$_2$Si$_2$.[1] Soon after the discovery of superconductivity in LaFeAsO$_{1-x}$F$_x$ at $T_c = 26$ K with the structure type ZrCuSiAs,[2] it was realized that the related compounds (AFe$_2$As$_2$ with A = Ba, Sr, Ca, Eu) in the ThCr$_2$Si$_2$ structure are also superconducting either with doping[3, 4, 5, 6, 7] or under pressure. [8, 9, 10]. While systems with Fe$_2$As$_2$ planes have the highest T_c's to date, superconductivity has been found in both structure types with either Ni$_2$P$_2$ (Refs. 11, 12) or Ni$_2$As$_2$ (Refs. 13, 14, 15, 16) layers.

Here we report the observation of superconductivity in single crystals of SrNi$_2$As$_2$ at T_c = 0.62 K, as determined by heat capacity, in the absence of a structural phase transition (below 400 K). Following our initial observation of superconductivity in BaNi$_2$As$_2$,[16] this represents the second superconducting system in the ThCr$_2$Si$_2$ structure with Ni$_2$As$_2$ layers. In addition, we report that our EuNi$_2$As$_2$ single crystals grown from Pb flux are not superconducting above 0.4 K.

Single crystals of SrNi$_2$As$_2$ and EuNi$_2$As$_2$ were grown in Pb flux in the ratio (Sr,Eu):Ni:As:Pb=1:2:2:20. The starting elements were placed in an alumina crucible and sealed under vacuum in a quartz ampoule. The ampoule was placed in a furnace and slowly heated 1050 °C, as described in Ref. 16. The sample was then cooled slowly (5°C hr^{-1}) to 600 °C, at which point the excess Pb flux was removed with the aid of a centrifuge. For SrNi$_2$As$_2$ the resulting plate-like crystals were heavily imbedded in a yet unidentified needle-like impurity phase. From single crystal x-ray refinements, the plate-like samples were confirmed to crystallize in the ThCr$_2$Si$_2$ tetragonal structure (space group no. 139, I4/mmm). The refinement for SrNi$_2$As$_2$ [R(I>2σ) = 3.7%] at 124 K yields lattice parameters $a = 4.1374(8)$ Å and $c = 10.188(4)$ Å and fully occupied (>98%) atomic positions Sr 2a(0,0,0), Ni 4d(0.5,0,0.25) and As 4e(0,0,z) with z = 0.3634(1) consistent with previous reports.[18, 19, 20] The refinement for EuNi$_2$As$_2$ [R(I>2σ) = 5.09%] at 124 K gives lattice parameters $a = 4.0964(6)$ Å, $c = 10.029(3)$ Å and fully occupied (>98%) atomic positions Eu 2a(0,0,0), Ni 4d(0.5,0,0.25) and As 4e(0,0,z) with z = 0.3674(2) also consistent with previous reports.[21, 22]

The in-plane electrical resistivity data for ANi$_2$As$_2$ (A=Ba, Sr, Eu) is shown in Fig. 1. All samples exhibit metallic behavior. SrNi$_2$As$_2$ is a relatively good metal with a RRR (= ρ(300 K)/ρ(4 K)) of 11, and a residual resistivity of 7 $\mu\Omega$-cm. The resistivity of EuNi$_2$As$_2$ exhibits

a kink at $T_m = 14$ K associated with magnetic ordering of the Eu^{2+} moments, consistent with previous reports.[22] For SrNi$_2$As$_2$, there is no evidence of a structural transition below 400 K , in contrast to BaNi$_2$As$_2$, which has a clear first order transition at $T_0 = 130$ K. The lack of a phase transition in SrNi$_2$As$_2$ is also provided by the heat capacity data shown in the inset of Fig. 3.[24]

Figure 2 presents the low temperature in-plane resistivity data for SrNi$_2$As$_2$ with fields applied parallel and perpendicular to the c-axis. In zero field, a sharp superconducting transition is observed at $T_c = 0.66$ K, defined as the mid-point resistive anomaly. With increasing magnetic field, the transition remains sharp and is quickly suppressed. The specific heat shown in Fig. 3 confirms the bulk nature of superconductivity in SrNi$_2$As$_2$. The zero resistance state coincides exactly with the onset of the specific heat transition, from which we extract a superconducting transition temperature of $T_c = 0.62$ K by an equal area construction. A fit to the data from 0.7 K to 3 K of $C/T = \gamma + \beta T^2 + \delta T^4$, a Sommerfeld coefficient $\gamma = 8.7$ mJ/mol-K^2 is obtained. Using this value, the ratio of the specific heat jump at T_c to the electronic specific heat is estimated to be $\Delta C/\gamma T_c \simeq 1.0$. From the β coefficient = 0.67 mJ/molK^4, one obtains a Debye temperature $\Theta_D = 244$ K.

The magnetic field-temperature $H-T$ phase diagram of SrNi$_2$As$_2$ is shown in Fig. 4 determined from the $\rho(T)$ curves in Fig. 2, along with the data for BaNi$_2$As$_2$ for comparison.[16] For SrNi$_2$As$_2$, the zero-temperature orbital critical field[25] $H_{c2}^*(0) = -0.7\,T_c\,dH_{c2}/dT_c$ is determined to be 210 Oe and 150 Oe for $H \parallel c$ and $H \parallel ab$, respectively. From this, the superconducting coherence length is estimated via $H_{c2}^*(0) = \Phi_0/2\pi\xi_0^2$,[26] yielding $\xi_0^{ab} = 1477$ Å, and $\xi_0^c = 1250$ Å. Values for the Fermi velocity $v_F^{ab} = 7.1 \times 10^6$ cm/s and $v_F^c = 6.0 \times 10^6$ cm/s are obtained from $\xi_0 = 0.18\hbar\,v_F/k_B\,T_c$. Surprisingly, while the absolute value of T_c in zero magnetic field is very similar for the two compounds, the anisotropy (H_{c2}^{ab}/H_{c2}^c) of the upper critical field, which is a factor of 2.1 in BaNi$_2$As$_2$, reverses sign (H_{c2}^c/H_{c2}^{ab}=1.4) in SrNi$_2$As$_2$ and the overall magnitude of the upper critical field is nearly an order of magnitude smaller. The differences in the H_{c2} between the two compounds may be due changes in electronic structure resulting from the structural transition in BaNi$_2$As$_2$ that is not present in SrNi$_2$As$_2$.

The specific heat, plotted as C/T, for EuNi$_2$As$_2$ is shown in Fig. 5 in magnetic fields up to 9 T $(H||c)$. A sharp anomaly occurs at the magnetic ordering temperature $T_m = 14$ K (consistent with the kink in $\rho(T)$, Fig. 1), as well as a broader hump at ~ 4 K. A magnetic

field along the c-axis modestly suppresses the transition, consistent with previous reports of antiferromagnetic ordering in polycrystalline samples.[22, 23] There is no indication of superconductivity above 0.4 K in $EuNi_2As_2$.

It is interesting that superconductivity with very similar transition temperatures is found both in $BaNi_2As_2$[16] and in $SrNi_2As_2$, despite the differences in structural parameters caused by the smaller Sr^{2+} ions, as well as the presence of a first order structural transition in $BaNi_2As_2$ that is possibly also magnetic. Recent theoretical work by Subedi and coworkers[28] indicate that the superconducting properties of the related Ni-analog LaNiPO may be explained within a conventional electron-phonon approach, yielding a low value of $T_c = 2.6$, consistent with experiment; the authors go on to suggest that the Fe-As superconductors may be in a separate class from their Ni-based counterparts. However, a scenario has been put forth by Cvetkivoc and Tesanovic[29] involving a multiband Fermi surface in the layered FeAs superconductors to produce the large values of T_c may also be an appropriate description of these ANi_2As_2 superconductors as well. Further work is in progress to elucidate the nature of the superconductivity in these Ni-based materials and its relation to fine details of the electronic structure.

In conclusion, specific heat and electrical resistivity measurements $SrNi_2As_2$ single crystals reveal bulk superconductivity at 0.62 K, which shows no sign of a structural/magnetic anomaly below 400 K. Magnetic ordering associated with the Eu magnetic moments is observed in single crystalline $EuNi_2As_2$. No evidence for superconductivity is observed in this compound above 0.4 K.

Acknowledgments

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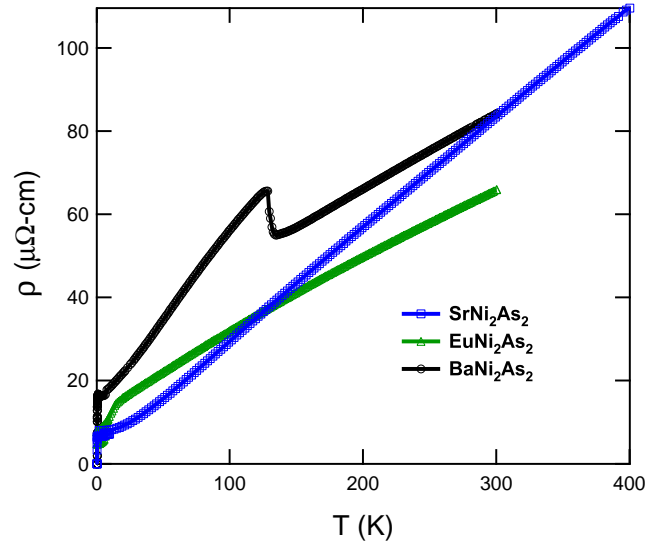[29] V. Cvetkovic and Z. Tesanovic, arXiv:0804.4678, (2008).

Figures

FIG. 1: (color online) In-plane electrical resistivity $\rho(T)$ (I∥ab) for selected ANi_2As_2 (A=Ba, Sr, Eu) compounds.
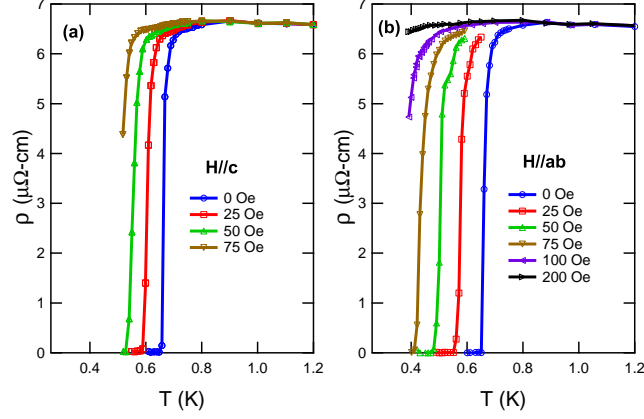
FIG. 2: (color online) Electrical resistivity $\rho(T)$ of SrNi$_2$As$_2$ showing the superconducting transition for $H \parallel c$ (a) and $H \parallel ab$ (b). The current was maintained perpendicular to the magnetic field.
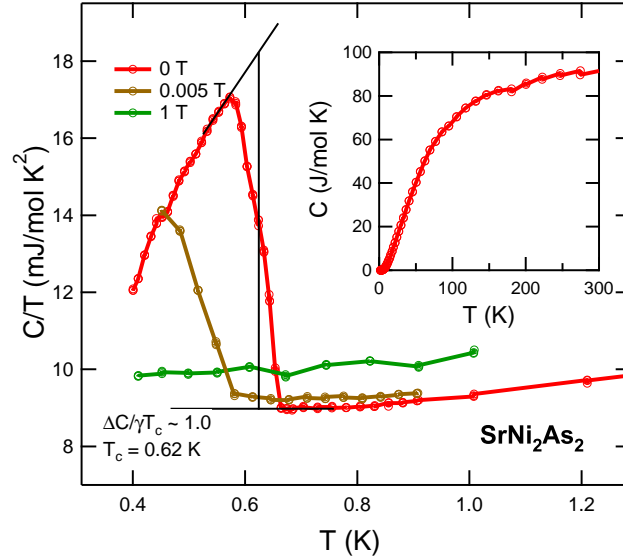

FIG. 3: (color online) Low temperature specific heat C versus temperature T of SrNi$_2$As$_2$ [24] for various magnetic fields ($H||c$). The inset displays the high temperature heat capacity with no indication of a first order phase transition at higher temperatures.
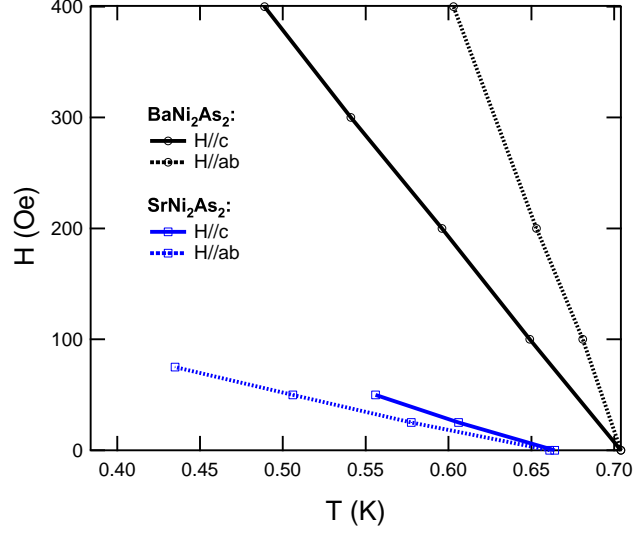
FIG. 4: (color online) Magnetic field-temperature $H - T$ phase diagram of $SrNi_2As_2$. The upper critical field H_{c2} for $H \parallel \hat{c}$ and $H \parallel \hat{ab}$ was determined by the resistive midpoint in Fig. 2(a) and (b). The data for $BaNi_2As_2$ from Ref. 16 is for the zero resistance state.
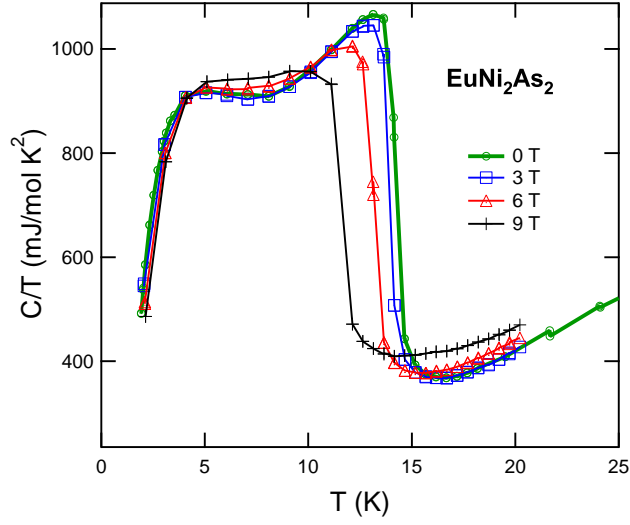

FIG. 5: (color online) Heat capacity data versus temperature for $EuNi_2As_2$ in zero and applied magnetic field [24]. The magnetic field was applied along the c-axis.